An Impurity-Doped Crystal Plate as an X-ray Monochromator

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Abstract
To increase the integrated reflexion intensity of a monochromating crystal, the strain distributed due to impurity introduction can be utilized and, compared with conventional mechanical processing, can be a more controllable method. From this viewpoint, numerical computations on the dynamical diffraction theory for distorted crystals have been performed for the 220 reflexion of Cu Kα X-rays by Si crystals with several different strain distributions; the one changing linearly with depth from $-10^{-3}$ at the surface to zero at 28 μm depth gives the maximum integrated intensity within the restriction that the full width of the intensity profile is 100°. The integrated intensity in this case is 5.3 and 4.5 times as strong as that of a perfect crystal plate for $\sigma$ and $\pi$ polarizations respectively. It is more enhanced for shorter wavelengths, e.g. nine times for Mo Kα. In addition, that intensity profiles for distributed strains are cut sharply at both ends is certainly favourable for the purpose of monochromating white X-rays. A preliminary experiment with an intentionally processed sample showed a fourfold intensity increase for a $\sigma$-polarized Cu Kα line with a full width of 80°.

1. Introduction
Strain distributions caused by impurities in silicon crystals have been analysed (Fukuhara & Takano, 1977a, 1977b; Fukuhara, Takano, Namba & Maki, 1980) from the intensity profile of X-ray Bragg reflexions, i.e. rocking curves. Computations based on a presumed strain distribution have shown good agreement with experimental data in some cases; these seem to suggest the possibility of artificial control of the intensity profile.

In addition, it was confirmed that the distribution of strain had a similar form to the distribution of the impurity with some limitations and the proportionality coefficients for boron and phosphorus in Si were determined experimentally. Thus, techniques such as impurity diffusion, growing crystals, annealing and etching developed extensively for silicon processing can be used to produce monochromator crystals specifically designed for diffraction experiments (Kohra, 1978).

The primary figure of merit for a monochromator is the integrated intensity, the integral of the intensity profile with respect to the angle of incidence, of the relevant reflexion. The strain due to impurity diffusion described above may possibly enhance integrated intensities in a more controllable way than conventional methods of mechanical surface processing.

Rustichelli and coworkers have studied diffraction characteristics of distorted crystals experimentally as well as theoretically in connexion with the monochromatization of synchrotron radiation and neutrons (Albertini et al., 1976; Boeuf et al., 1978; Boeuf, Melone, Puliti & Rustichelli, 1978). They were interested mostly in the effect of bending on monochromator crystals; to achieve an appreciable gain a very short bending radius (≤ 1 m) was needed while the radius should be consistent with other conditions for focusing. Thus the present report is confined to the effect of distortion due to impurity doping on the integrated intensity enhancement.

The following part of this report is devoted to the theoretical deduction of a strain distribution corresponding to a maximum integrated intensity for the Si 220 reflexion and finally gives preliminary experimental data from a sample crystal processed for this purpose. The theoretically optimum strain distribution may exceed the limit of conventional silicon processing, but it still serves as an ideal target and will show the direction of best endeavours.

2. Theoretical estimates
For the sake of simplicity and clarity of discussion, the following assumptions are postulated: the crystal plate is thick enough that only expansion or contraction in depth can exist as strain, and the relevant reflexion is of symmetric Bragg type. Numerical computations are based on dynamical diffraction theory for distorted crystals (Takagi, 1962, 1969; Taupin, 1964) and are described elsewhere (Fukuhara & Takano, 1977). Since we are interested in a strong reflexion of Si, the 220 reflexion is used. The wavelength is fixed at that of Cu Kα X-rays and the polarization is of $\sigma$ type for most of the computations unless otherwise stated.

The following points should be noted before we describe the numerical computations used to find the maximum integrated intensity. Firstly, the full width of the intensity profile is roughly proportional to the maximum value of the strain in the crystal plate. The latter is restricted by the upper limit of the impurity concentration in silicon or by the appearance of...
dislocations; supposedly, maximum full width cannot exceed 100′ for Cu Kα, which corresponds to an absolute value of strain or 1 × 10⁻³. Secondly, the mean absorption effect determines the upper limit of the layer thickness effective in X-ray reflexion irrespective of the strain distribution; thus the layer thickness is about 30 μm for Cu Kα. These two points give us a rough idea of the desirable strain gradient and reduce our search area. In Table 1 are tabulated parameters used in the computations and important quantities for the reflexion. The ratio R of the computed integrated intensity to that of a perfect crystal plate will be shown as the measure of the intensity enhancement for each strain distribution.

Next let us consider strain distributions having the form of a flight of equal steps, for which the total height and the total horizontal width are denoted by e₀ and zₚ respectively. The intensity profile of a flight of five steps is shown in Fig. 1 as an example. The smallest peak on the left comes from the reflexion at locations deeper than zₚ. The strain difference between adjacent steps is so large that peaks are clearly separated in their angle of incidence; however, the integrated intensity ratio R = 3.5 here. To fill up the spaces, a flight form of more steps has been tried as shown in Fig. 2, which gives R equal to 5.0.

Fig. 3 is for the same strain distribution as in Fig. 2 except for the sign of the strain; i.e. the crystal plate is expanded in depth near the surface. The reduction of R compared with Fig. 2 is explained by consideration of the Borrmann effect; in the contraction case the Bragg reflexions in the deeper layers are enhanced by the anomalous transmission related to the Bragg reflexion in upper layers, while it is suppressed in the expansion case by the anomalous absorption in the upper layers. It can thus be concluded in general that the strain should be of a contraction type for our purpose.

At the limit of many steps with the same e₀ and zₚ, the distribution has a linear form to depth zₚ but it gives almost the same value of R as in Fig. 2. For a fixed value of e₀ the maximum value of R has been found to be 5.3 at zₚ = 28 μm (Fig. 4); for a larger zₚ, the intensity profile is weakened on the left side which eventually reduces R. The value of R still increases slowly for a larger absolute value of e₀, which, however, does not seem to be practical, as mentioned before.

As is well known, the effective scattering factor is reduced by a factor cos 2θB for π polarization, θB being the Bragg angle. This effect is common to both a perfect

Table 1. Parameters used in the computations

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<th>Cu Kα</th>
<th>Mo Kα</th>
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<tr>
<td>ϕ₀</td>
<td>-1.51 × 10⁻⁵ + 3.83 × 10⁻⁷i</td>
<td>-3.18 × 10⁻⁶ - 2.03 × 10⁻⁸i</td>
</tr>
<tr>
<td>ϕ₁₂₀</td>
<td>9.28 × 10⁻⁴ + 3.71 × 10⁻⁷i</td>
<td>1.94 × 10⁻⁶ + 1.97 × 10⁻⁸i</td>
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<td>μ₀⁻¹ (μm)</td>
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<td>1.08</td>
</tr>
<tr>
<td>μ₀⁻¹ (μm)</td>
<td>25.7</td>
<td>103</td>
</tr>
</tbody>
</table>

Fig. 1. The intensity profile of Cu Kα Si 220 (σ polarization) for a strain distribution e₀ = -10⁻³, zₚ = 23 μm, 5 steps. The abscissa shows the angular deviation of incidence from the Bragg position for locations deeper than zₚ, R = 3.5.

Fig. 2. The intensity profile of Cu Kα Si 200 (σ polarization) for a strain distribution e₀ = -10⁻³, zₚ = 23 μm, 15 steps. R = 5.0.

Fig. 3. The intensity profile of Cu Kα Si 220 (σ polarization) for a strain distribution e₀ = +10⁻³, zₚ = 23 μm, 15 steps. R = 4.0.
crystal and strained crystal plates but diminishes the anomalous transmission in the upper layers as previously described in the contraction case; thus, the $R$ value for negative $e_0$ should be smaller for $\pi$ polarization than for $\sigma$ polarization. For $\pi$ polarization, $R$ is estimated as 4.4 for the strain distribution of Fig. 4.

The ratio $R$ for the same strain but shorter wavelength, that of Mo $K\alpha$, is 9.1, which is much larger than for Cu $K\alpha$ owing to the weaker absorption effect (Fig. 5).

3. Concluding remarks

It should be noted that the favourable strain gradient is similar at any wavelength for a symmetric Bragg reflexion; by the favourable gradient we mean that for which the intensity profile does not consist of separate peaks (cf. Fig. 1) but has a solid shape (cf. Fig. 2). This can be deduced from the dynamical theory applied to distribution forms of a flight of steps; for a length $\Delta z$ of a step roughly equal to the extinction depth, the height of the step $\Delta e$ should be the value of strain so as to shift the Bragg position by the intrinsic full width of the Bragg peak. Thus, a simple calculation leads to

$$\frac{\Delta e}{\Delta z} = \frac{(\varphi_s)^2}{\lambda^3} \sin \theta_B \times \text{(numerical factor)}$$

Fig. 4. The intensity profile of Cu $K\alpha$ Si 220 ($\sigma$ polarization) for a strain distribution $e_0 = -10^{-3}$, $z_s = 28 \mu m$, linear type. $R = 5.3$.

Fig. 5. The intensity profile of Mo $K\alpha$ Si 220 ($\sigma$ polarization) for a strain distribution $e_0 = -10^{-3}$, $z_s = 28 \mu m$, linear type. $R = 9.1$.

where $\lambda$ and $\theta_B$ are the relevant wavelength and the Bragg angle, respectively. For a fixed Bragg reflexion, the magnitude of $\varphi_s$ is roughly proportional to $\lambda^2$ except near the absorption edge and $\lambda/\sin \theta_B$ is a constant so that $\Delta e/\Delta z$ should be roughly independent of $\lambda$.

The computed intensity profiles for single crystals with strains are cut sharply at both ends in contrast to the Gaussian tails expected for random mosaics of crystallites. This fact is of practical advantage in monochromating white X-rays.

For the full width of the profile to be 100", we conclude that the strain distribution with $e_0 = -10^{-3}$, $z_s = 28 \mu m$ gives an optimum integrated intensity for the Si 220 reflexion of Cu $K\alpha$ X-rays and that the fine details of the distribution form are not critical. For different profile widths, $e_0$ should be changed in proportion to the width but $z_s$ must be almost constant for the same wavelength.

Experimental study along these lines is also being carried out by the present authors. The result of preliminary work is shown in Fig. 6. The sample was grown epitaxially by a chemical vapour-deposition method in which the boron concentration was intentionally changed during the growth. The rocking curve was recorded with a triple-crystal diffractometer on which the first crystal was set for the 511 reflexion to select only $\sigma$ polarization. The other features of the experimental arrangements were similar to those described previously (Fukuhara & Takano, 1977a). The rocking curve revealed that the surface strain was small but the distribution shape was increased and the intensity ratio $R$ was 4.0. Detailed experimental data will be published in the near future.

References


Fig. 6. The experimental rocking curves of Cu $K\alpha$ Si 220 ($\sigma$ polarization) from a perfect crystal and from a sample with a boron-doped epitaxic layer: for the latter the surface concentration is estimated at $1.2 \times 10^{26}$ atoms/m$^3$, $z_s$ is $28 \mu m$ and $R = 4.0$. 


